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Using Laser-Induced Incandescence to Measure Soot/Smoke Concentrations

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USING LASER-INDUCED INCANDESCENCE TO MEASURE SOOT/SMOKE CONCENTRATIONS

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INTRODUCTION

The production of particulates, notably soot, during combustion has both positive and negative ramifications. Exhaust from diesel engines under load (for example, shifting gears), flickering candle flames and fireplaces all produce soot leaving a flame. From an efficiency standpoint, emission of soot from engines, furnaces or even a simple flickering candle flame represents a loss of useful energy.

The emission of soot from engines, power generation facilities and incinerators is a serious environmental pollutant and poses a health risk. Yet other industries intentionally produce soot as carbon black for use in inks, copier toner, tires and as pigments. Similarly, the presence of soot within flames can act both positively and negatively. Energy transfer from a combustion process is greatly facilitated by the radiative heat transfer from soot yet radiative heat transfer also facilitates the spread of unwanted fires. To understand soot formation and develop control strategies for soot emission/formation, measurements of soot concentration in both practical devices such as engines and controlled laboratory flames are necessary.

Incandescence is a familiar process due to its role in light generation. Incandescent light bulbs emit light when the carbon coated tungsten filament is heated by the passage of an electric current. While all objects emit visible radiation as described by Planck's law, radiant intensity is invisible to the unaided eye for object temperatures below 900 K. If objects are heated to temperatures exceeding 3000 K, however, all visible wavelengths are emitted with sufficient intensity that the object appears "white-hot." The electromagnetic radiation intensity increases as the temperature of the objects increase, with the peak wavelength of the emission shifting towards smaller wavelengths. For example, within a flame, the soot temperature is elevated resulting in an increase in the intensity of visible (usually yellow) and nonvisible radiation.

As a diagnostic to determine soot concentration or soot volume fraction (defined as the volume of soot per unit volume of space, a dimensionless quantity), the phenomenon of incandescence can be used. When pulsed high intensity laser light further heats soot to temperatures far above flame temperature, the soot will incandesce strongly throughout the visible region of the spectrum. The fact that pulsed high intensity laser light can elevate the soot particle temperature even when the soot glows in flames was observed long ago but considered an interference in pulsed laser diagnostics (1). Only recently have researchers demonstrated that this laser-induced emission can be used as a measure of the soot concentration (2-6), thereby validating theoretical predictions (7-11). In accord with the Planck radiation law, soot's radiative emission at these elevated temperatures increases in intensity and shifts to blue wavelengths as compared with the nonlaserheated soot and flame gases (12). Because it is pulsed and blue shifted, the LII signal is readily isolated from natural flame emission. This feature is illustrated in the lower panel of figure 1.

Alternative methods of measuring soot volume fraction include physical sampling of the soot and measuring the absorption caused by soot. Sampling lacks high temporal and spatial resolution and requires an intrusive probe that can perturb the combustion process. Absorption lacks high temporal resolution, can be complicated by scattering and requires knowledge of the path length and assumptions regarding soot physical properties. In contrast, LII, offers high spatial and temporal resolution and can be used for multidimensional measurements (3-6, 13-16). Additionally, LII provides a measure of the soot volume fraction nearly independent of unknown contributions from both scattering by soot aggregates and absorption by polycyclic aromatic hydrocarbons.

In addition to its application to steady-state systems, such as laminar gas-jet diffusion flames, LII is also ideally suited to measuring soot concentrations in phenomena with rapid timescales lacking spatial symmetry such as turbulent combustion processes (4, 17). This is possible because measurements may be obtained from a single laser pulse using detection times as short as 10 ns. Since LII is not a line-of-sight technique, it also possesses geometric versatility enabling its application to nonaxisymmetric systems.
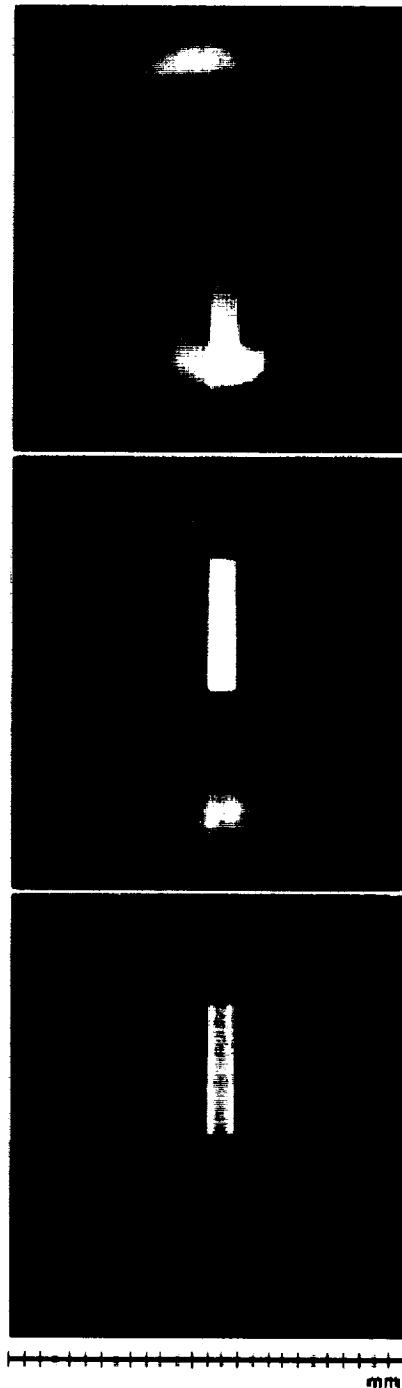
Figure 1.—Color Photograph of an acetylene-air diffusion flame through a viewing port within a chimney. The LII images (Middle and lower panels) were produced by the passage of a laser sheet though the flame midsection. The white appearance of the LII signal (middle panel) illustrates the spectrally broad emission produced by the laser heated soot. The bottom LII image was produced with a blue transmitting glass filter placed in the signal optical path. The increase in the short-wavelength emission from the laser heated soot relative to the nonlaser-heated soot and flame gases is readily apparent.

THEORY AND VALIDATION

Several theoretical analyses describe the interaction of laser radiation with suspended particles such as soot (1, 4, 8-11). For submicrosecond laser pulses, energy balance equations indicate that the energy addition rate by the laser greatly exceeds the loss rate by thermal conduction, vaporization, or radiation. For example, the temperature of a soot particle rapidly rises to the vaporization temperature of carbon, approximately 4000 K, for laser intensities of 1×10^7 W/cm^2 or greater. Equilibration of the absorbed energy within the particle occurs rapidly, on the time scale of the laser pulse (e.g. ns), but heating of the medium surrounding the particle occurs on a longer time scale (e.g. usec). For particles in the Rayleigh size regime ($\pi d/\lambda < 0.1$, where d is the particle diameter and λ is the relevant wavelength) absorption and emission of electromagnetic radiation occurs volumetrically (18).

Because the primary particles composing a soot aggregate are typically 20 to 50 nm, this condition is readily fulfilled in both laboratory flames and most commercial combustion processes. To the extent that the primary particles act as point-contacting, noninteracting spheres in the aggregate, as is commonly assumed, the physical dimensions of the aggregate will not affect the absorption or emission process as indicated by theoretical calculations. Thus, theory predicts that incandescence is a measure of soot volume fraction, which is the attribute of soot that governs many physical processes such as radiative heat transfer and soot emission from flames.

Measurements show that the LII signal is linearly proportional to the soot volume fraction (2, 3, 5). Unlike the absorption method, LII yields a relative measure of soot concentration that requires calibration to extract quantifiable soot concentrations (2, 3, 5, 14-17). In principle, quantification could be obtained by detailed theoretical modelling of the signal. At present, modelling the transient heating and cooling of the soot along with time-varying structural transformations of the heated soot poses a formidable challenge. Currently, calibration of the technique for absolute soot concentration may be made by in-situ comparison of the LII signal to a system with a known soot volume fraction determined through traditional methods. Using this empirical calibration procedure, LII has been used to measure soot volume fraction in steady-state (2, 3, 5, 6, 13-16) and time-varying diffusion flames (4, 5, 15-17), premixed flames (3, 19) and within engines (12, 20, 21).

APPARATUS

Figure 2 illustrates the typical apparatus for performing LII measurements of soot concentration. Light from a pulsed laser is directed through various beam delivery optics to the combustion process. Nd:YAG and pulsed dye lasers are most commonly used for LII measurements given their widespread availability and ease of use. Since focused Q-switched laser pulses can readily reach intensities greater than 1×10^7 W/cm^2, modestly priced solid-state lasers with low energy can be used for "point" or line measurements. For these measurements, a monochromator and time-gated detection of a photomultiplier tube signal can provide spectral and temporal rejection of natural flame luminosity while providing spectrally and temporally selective detection of the LII signal, which is spectrally broadband and varies in time.

For one- or two-dimensional LII measurements, additional beam expansion and/or sheet forming optics are required to manipulate the spatial dimensions of the laser beam. For these measurements, a gated intensified array camera is required. A bandpass interference filter and the gated camera intensifier provide for both rejection of natural flame emission while enabling spectrally and temporally precise detection of the LII signal. Synchronization electronics are generally required to coordinate in time the laser pulse and gated integrator or camera intensifier gate pulse. With either method of detection, a computer is commonly used to store and subsequently process the data.

CHARACTERIZATION

Figure 3 illustrates the spectral and temporal characteristics of the LII signal. Figure 3(a) shows the broad wavelength distribution of the LII signal (typical of incandescence). The emission curve, corrected for the instrument response, was obtained by spectrally resolving the LII signal at its peak intensity using a scanning monochromator. As expected for an object less than 4000 K, the emission in the visible spectral region increases with increasing wavelength because the peak emission wavelength lies at longer wavelengths. Often the signal is detected in the near ultraviolet because the quantum efficiency of photocathode materials peaks in this region and natural flame luminosity is far lower in intensity than at longer wavelengths.
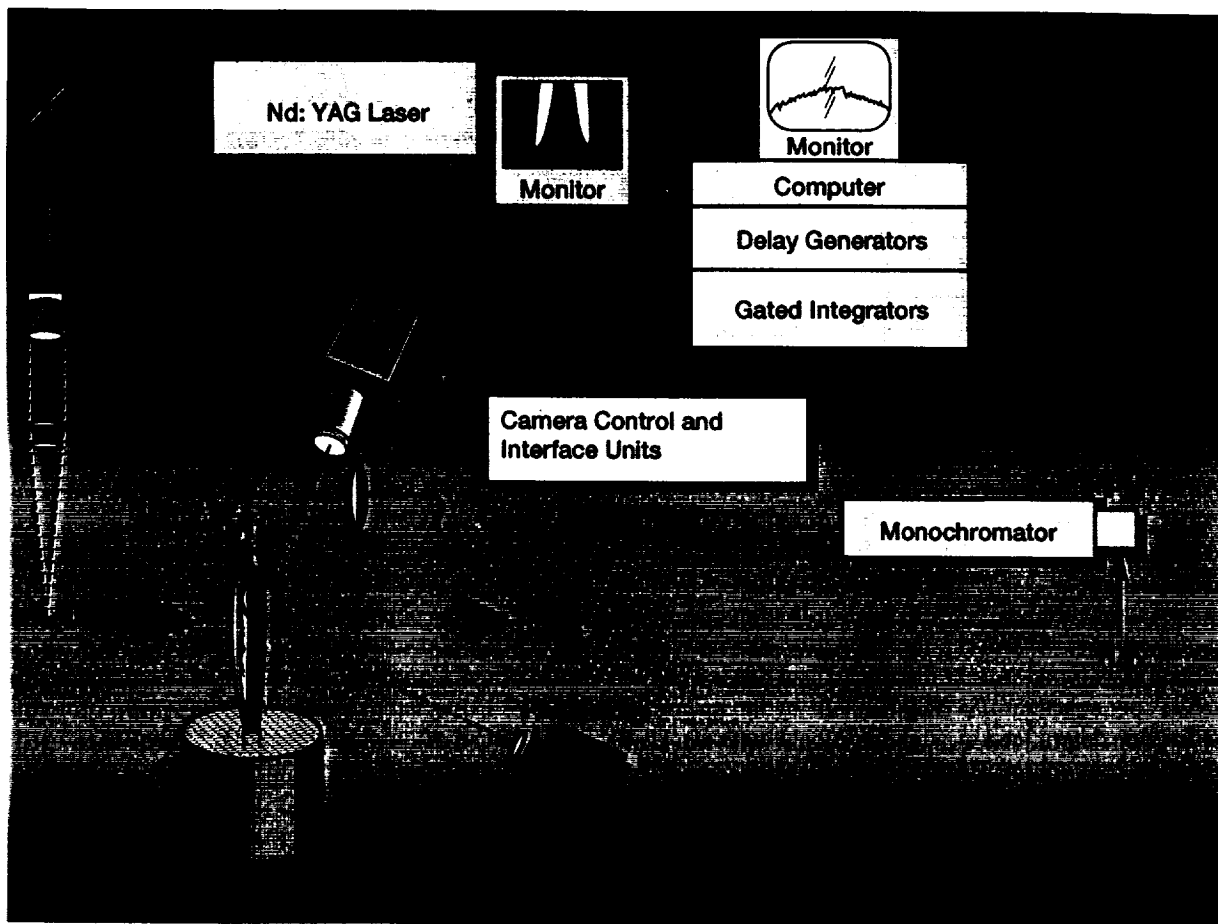
Figure 2.—Experimental schematic of typical apparatus for LII measurements. Typical apparatus for LII measurements includes a pulsed laser, beam-delivery optics, low-light detectors such as a photomultiplier tube and/or gated intensified array camera, and timing and signal acquistion electronics.
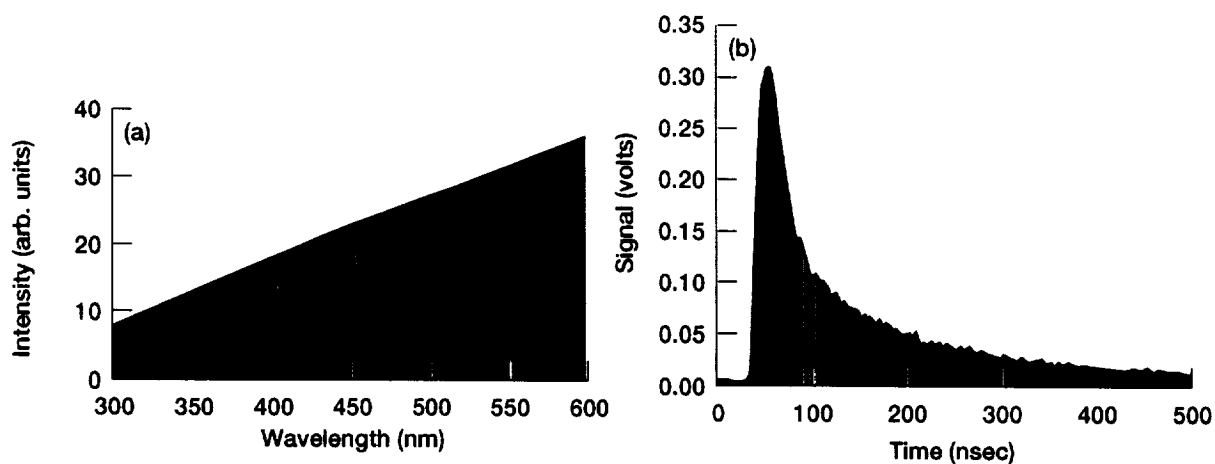

Figure 3.—(a) Spectrally resolved emission produced by the incandescing soot as a result of the pulsed laser heating. (b) Temporally resolved LII signal. The arrows indicate a commonly used spectral region for detecting LII.

Attempts to fit such spectra to black-body curves however have met with limited success. This likely reflects the variation of particle temperatures due to both the variation in laser intensity across the laser beam profile and the distribution of soot primary particles sizes. The utility of such information is that laser-induced spectral interferences can readily be identified. These interferences arise from electronically excited small molecules or radicals produced by the high intensity laser light which can fluoresce in the wavelength interval in which the LII signal is collected. Such signals are experimental artifacts, not necessarily representative of the soot concentration and should be avoided in detecting the LII signal (3, 16, 22).

Figure 3(b) illustrates the temporal variation of the signal. The emission from the laser-heated soot rapidly rises, reaching a peak and subsequently decays after the excitation laser pulse. The decay reflects the cooling of the soot via conductive and convective heat transfer processes (7-11). Empirical studies have shown that the best measure of soot concentration is obtained by detecting the signal during and shortly (within a few tens of nanoseconds) after the excitation laser pulse (11, 22). During this time period, different cooling rates associated with different primary particle sizes have the least effect on the particle temperature and consequently upon the LII signal.

Of course, prompt temporal detection of the signal is only accurate provided that spectral interferences can be avoided by detecting other wavelengths to serve as a measure of the incandescence intensity. If not, delayed detection of the LII signal can be used since fluorescence interferences are generally short-lived relative to the LII signal. It is best to avoid such interferences by the use of long wavelength excitation. At longer excitation wavelengths, fewer electronic resonances exist to produce electronically excited molecules and radicals that may fluoresce (23).

EXAMPLES

Knowledge of the soot volume fraction and spatial distribution is central to understanding several types of combustion phenomena. As an illustration of the spatial and geometric capabilities of the technique, figure 4 shows LII images of a gas-jet diffusion flame of ethylene surrounded by an air coflow. The image on the left was obtained by forming the laser beam into a vertically-oriented sheet and passing it through the center of the flame. This gives the soot distribution for a "slice" through the flame with a thickness corresponding to that of the laser sheet. The ring shaped images on the right were obtained by orienting the laser sheet in a horizontal plane passing through the flame at heights of 10.0, 14.1 and 16.9 mm above the burner nozzle (as indicated by the arrows) and imaging the laser-induced incandescence from above the non-soot emitting flame. The ruler at the left gives the spatial scale in millimeters.

These images show how the distribution of soot changes from a thin annular shell near the base of the flame which thickens and eventually fills in at higher heights above the nozzle. These results are consistent with previous measurements in similar flames using line-of-sight extinction methods. The latter technique requires a series of sequential measurements or multiple simultaneous measurements followed by deconvolution algorithms. In contrast, each of the LII images was obtained using a single laser-shot with a signal collection time of 100 nsec for each image.

Perhaps the best illustration of the spatial and temporal capabilities of LII is its application to timevarying systems such as those that occur in turbulent combustion. Turbulent diffusion flames are widely used in practical and commercial devices so knowledge of soot concentration is vital to assessing and improving the performance of these devices. The left panel of figure 5 shows a LII image from a turbulent ethylene/air diffusion flame of Reynolds number 2500 produced by a single laser pulse. The instantaneous spatial distribution of soot concentration provided by such an image is unavailable by any other technique. Images such as these are also illustrative of the type of information that can be collected from measurements within engines. With calibration, the intensity contours of the image shown in the right panel of figure 5 can be assigned absolute soot concentration values. Such information presents a step towards validating soot formation models within turbulent flames and validating radiative heat transfer from such flames.

Knowledge of the soot concentration in combustion exhaust gases is important for developing and assessing control strategies. Laboratory studies often involve measuring changes in emitted soot concentration resulting from the various burning conditions that can occur within engines under varying loads. Real time measurements of soot concentration or extent of mixing in the post-combustion exhaust are desirable in these applications. LII offers the possibility of real-time spatially resolved measurements of soot concentration within either exhaust gases or a process stream.
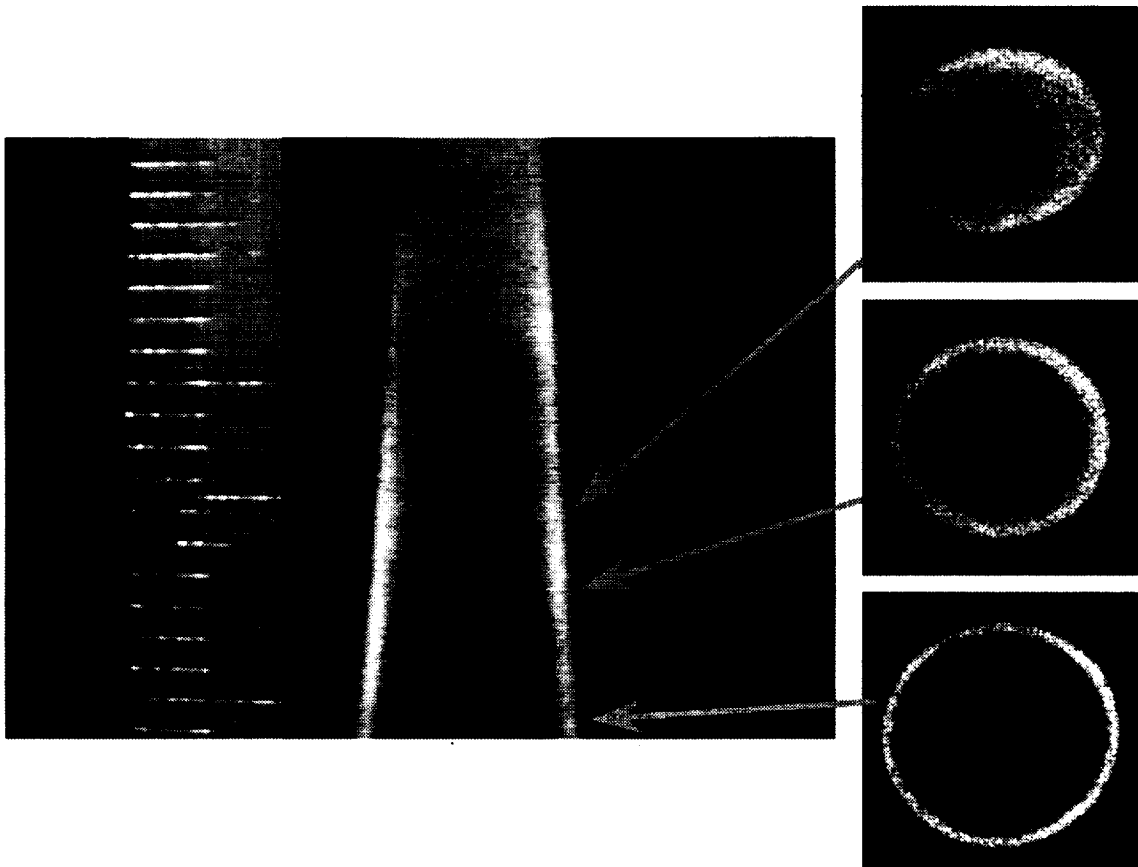
Figure 4.—LII images obtained with single laser pulses within a laminar (steady-state) gas-jet diffusion flame of ethylene. The spatial scale is in millimeters.
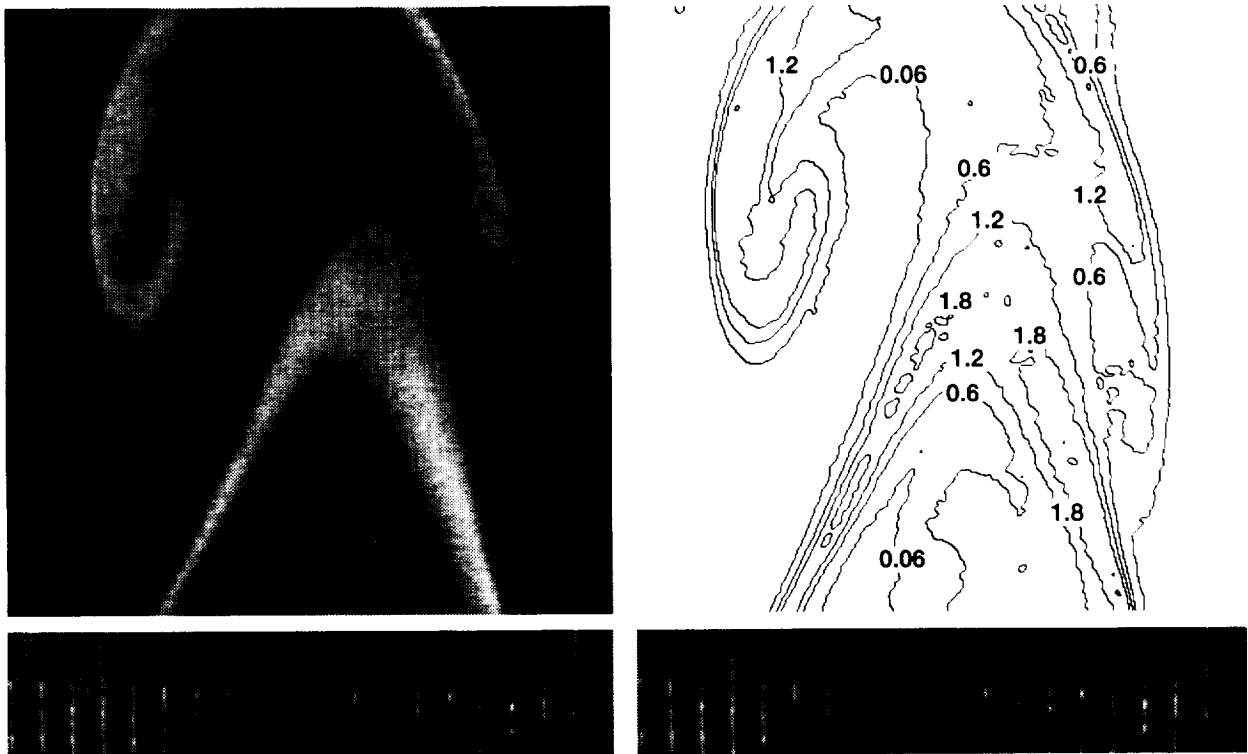
Figure 5.—LII image and associated intensity contour plot of soot within a turbulent gas-jet diffusion flame of ethylene produced by a single laser pulse. The spatial scale is is millimeters.

Figure 6.—Images through a viewing port into a
chimney containing a soot-laden exhaust stream.
The top panel shows a ruler giving the spatial scale
in inches. The middle and lower panels are LII
images produced with one and two single laser
pulses, respectively, of soot within the post com-
bustion exhaust.

To assess the applicability of LII to these systems, a soot containing exhaust stream was created in a chimney. This chimney was topped by a glass cylinder with 4 windows to provide optical access. Light at 1064 nm from a pulsed Nd:YAG laser initiated the incandescence emission from the soot. Figure 6 shows LII images that reveal the soot distribution within the chimney. The middle and lower panels are LII images while the top panel shows a ruler with a scale in inches which was inserted into the image plane prior to performing experiments. An experimental advantage of exhaust measurements is the lack of flame luminosity thereby relaxing wavelength discrimination requirements on signal detection apparatus and enabling the use of simple detectors. Because the incandescence was visible-to-the-eye, an ordinary 35 mm film camera was used with black and white film to obtain the LII images. The middle panel was produced by a single laser pulse while the lower panel shows two overlayed LII images obtained by keeping the camera shutter open for a time duration overlapping 2 laser pulses. The detection of the signal using an ordinary 35 mm film camera attests to the high signal levels offered by the technique.

As a diagnostic for soot concentration, LII has recently emerged as the method of choice in a wide variety of laboratory investigations. Quantitative data obtained by advanced diagnostics are often needed to evaluate the performance of combustors, improve their design and/or operation and provide new insights into combustion science. Optical diagnostics are well-suited towards these measurements given their non-intrusive nature. With additional characterization and improved understanding of the technique, LII offers great potential in commercial combustion applications.

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Laser-induced incandescence offers great advantages in measuring soot concentrations. A brief summary of the technique and some illustrations of its capabilities is presented here.

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